DETERMINATION OF METASTABLE POPULATIONS IN AN ARGON GLOW DISCHARGE

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Lieutenant, United States Navy

Submitted in partial fulfillment of
the requirements for the degree of

MASTER OF SCIENCE
IN
PHYSICS

United States Naval Postgraduate School
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ABSTRACT

The absorption of a beam of 8115A radiation by a striated Argon glow discharge was examined to determine the population density of metastable excited atoms in the discharge. Difficulty in obtaining an irradiating beam of sufficient intensity precluded reportable results. The re-emission characteristics of the same discharge were examined extensively for the same reason. Lack of reportable results is attributed to the lack of an illuminating beam of sufficient intensity.

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1. INTRODUCTION

1.1 Earlier Theoretical and Experimental Work

Since first reliably reported in the 1920's,[1] moving striations have been the object of no small amount of investigation and theorization.

Early investigations of striations concerned themselves with measurements of frequency, velocity and wavelength. The rotating mirror and photo-cells were used as the primary investigative tools and the results were compared with oscillograms of tube potential and tube current.

The really first significant attempt to probe the mysteries of moving striations was reported by Pupp in 1933 and 1934.[2] Pupp concluded that moving striations and anode oscillations were independent phenomena and that moving striations were produced within the positive column. Of far greater import, however, was Pupp's technique of preventing anode oscillations by setting up an auxiliary discharge at the anode.

In 1949 Loeb[3] offered the suggestion that the cathode was the seat of oscillations which might initiate striations. W. P. Allis theorized in 1950[4] that moving striations were caused by a buildup of metastable atoms in the anode spots followed by an increased ionization rate and a space charge layer which moved away from the anode.

Also in 1950 Fenty[5] reported on his studies of the metastable $^3P_2$ level in Mercury. He explained the increased

tube potential, the increased 2537A intensity and suppressed moving striations as results of destroying metastable atoms with resonant radiation. This suggests that striations in unbuffered Mercury discharges are associated with a two-step ionization process. Such a suggestion was formalized by Donohue and Dieke in 1951 (6) to explain a 20 μs phase lag in excitation of the 2537A and 4358A lines in moving striations in Mercury.

In this same paper Donohue and Dieke (6) concluded that negative striations originate in the negative glow region near the cathode and are triggered by approaching positive striations. The positive striations were supposed to have been caused by a buildup of a cloud of ions near the anode which was released simultaneously with the occurance of a maximum of tube potential. Perhaps most important from this only slightly historical vantage point was the belief of Donohue and Dieke that moving striations and oscillations were normally present in the positive column of the inert gas and Mercury discharges (within limited ranges of currents and pressures) and that lack of them was to be viewed as an exception to the rule. They further suggest that moving striations have a principal role in sustaining the glow discharge. In papers published in 1951 and 1952 (7) Zaitsev concluded that, contrary to Pupp, anode oscillations and moving striations were not necessarily independent phenomena but that anode oscillations could establish striations. He further

offered the suggestion that any oscillation might initiate moving striations and that oscillations occurred naturally in the cathode and anode ends of the positive column.

In 1952 Gordeev[8] postulated that both positive and negative striations were a direct result of electron oscillations. The positive striations were said to be initiated by electrons being accelerated in the anode fall region while negative striations were reflections of the positive striations off the cathode or cathode glow.

Meissner and Miller[9] in 1953 published the results of their investigations in He, Ne, A and Xe. The effect of resonant radiation on tube potential in these gases corroborates the effects noted earlier by Penty[5] in Mercury. In 1954 Pekarek[10] reported, in one of a long series of papers, the proposal that there exists in a positive column "waves of stratification" which originate in the cathode region. These, he says, are links in an interesting feedback mechanism which connects striations, anode oscillations, waves of stratification and cathode disturbances. In another of his series of papers[11] he adds evidence to this thesis.

In 1955 Watanabe and Oleson[13] undertook to examine the positive column theoretically. They concluded that travelling waves of positive ion and electron density can exist in the column. They are careful not to identify these as striations. Robertson[14] presented in 1957 his theory of moving striations, the theory which prompted this author's

investigation. Robertson's theory is the first theoretical mathematical treatment which specifically includes the metastable states. Robertson bases the treatment on continuity considerations for positive ions, electrons and metastable atoms. He considers a longitudinally uniform, axially symmetric plasma which can be excited by some outside source. He shows that for the spatially uniform plasma a large concentration of metastable atoms is needed for instability to appear. Further, when variations in metastable concentrations are ignored, travelling charge density waves, similar to those of Watanabe and Oleson,[13] can be predicted and when all diffusion is ignored travelling density waves moving in either direction can be predicted. The direction of travel depends upon production and loss processes. This last is in consonance with the results of Rademacher and Wojaczek[15] who show also that a disturbance in the positive column propagates more readily toward anode than cathode. In 1958 in a further paper of his series Pekarek[12] concludes that negative striations involve a single step ionization process while positive striations are tied-up with multi-step ionization processes. This latter seems to support the earlier suggestions of Donohue and Dieke.[6]

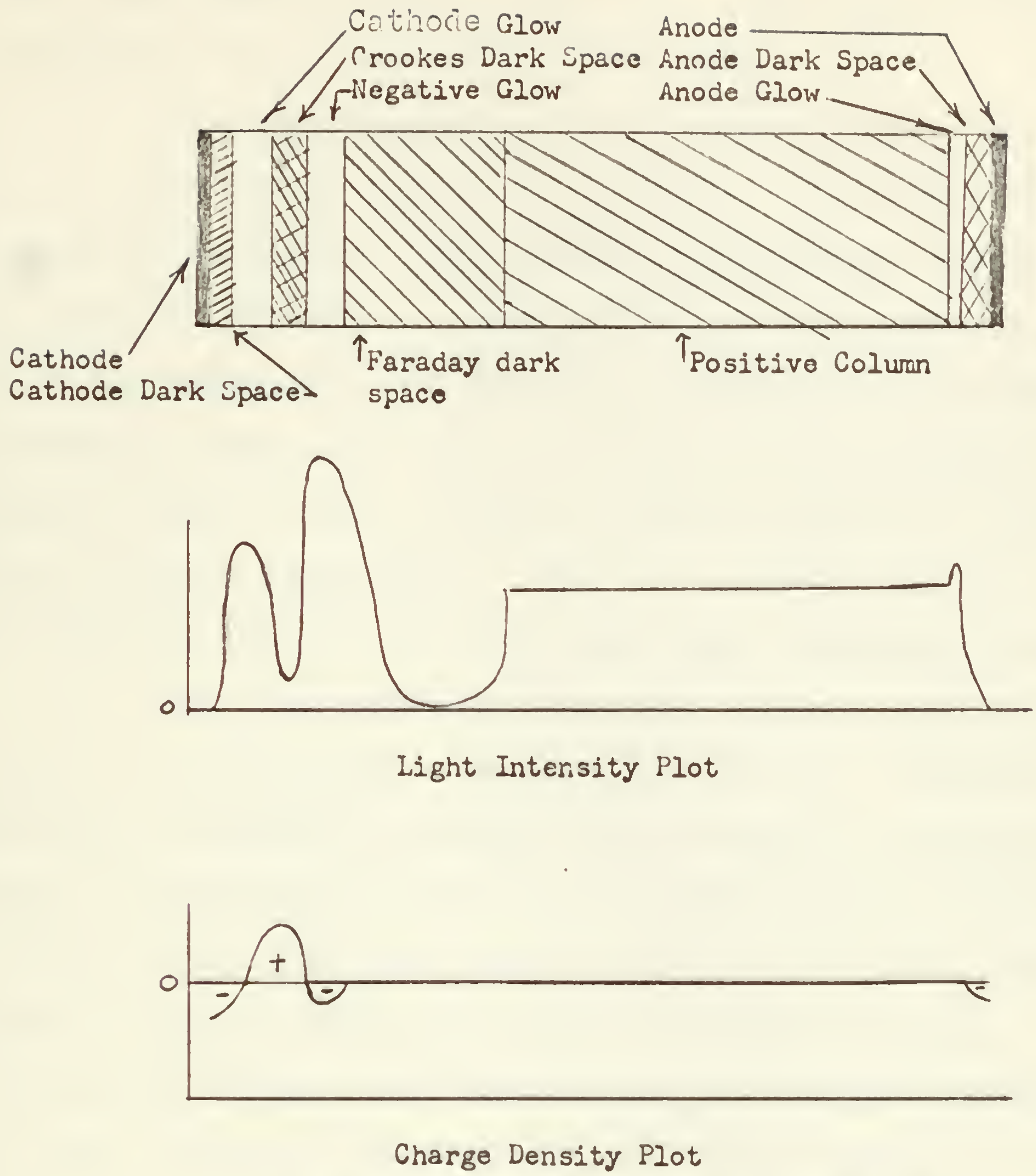
In 1960 Cooper[16] reported that moving striations could be synchronized with and seemingly driven by external oscillations over a range of frequencies. Hakeem and Robertson[17] in 1960 and again in 1961[18] reported on their work

in the alkali vapors. The results, viewed against the background of knowledge to that date, led them to conclude in the latter paper that the existence of striations requires the existence of metastable levels and that in view of this Robertson's earlier theory[14] could be profitably polished.

It should be noted that through the years many authors have attempted to explain moving striations from a cause and effect point of view which has led the mass of them to consider some sort of oscillation or perturbation of the positive column as the initiator of striations. These treatments, if carried to the limit of steady state moving striations eventually require repetitive plasma perturbations which do not have firm grounding in experiment. Before jumping to the Robertson theory which has oscillations as a result rather than a requirement it will be well to mention that the plasma oscillations are still being studied extensively. Significant results are sparse but encouraging as evidenced by two recent reports from Stanford University[19-20] which show good correlation between theory and experiment for R.F. oscillations in the plasma sheath.

1.2 THE GLOW DISCHARGE

The idealized and much to be desired glow discharge is shown in Fig. 1. Glow discharges characteristically occur when discharge pressures are below a few centimeters Mercury but can be sustained at lower pressures. Depending upon the tube length and applied potential difference, there is a pressure below which the positive column ceases to exist



A GLOW DISCHARGE

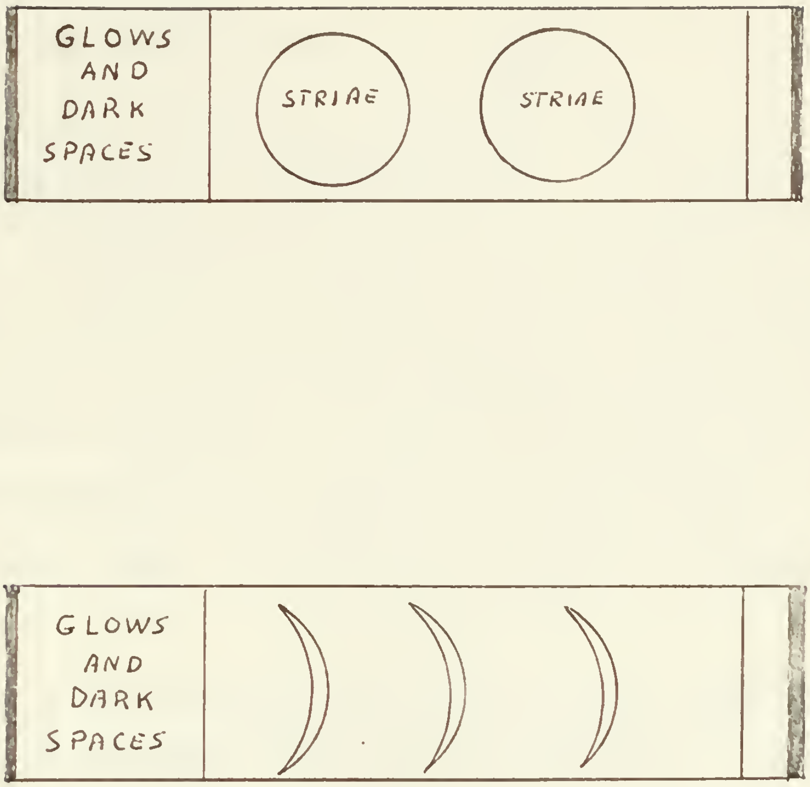
FIGURE - 1

as such and the negative glow and Faraday dark space fill nearly the entire volume of the tube. Such a low pressure discharge is of no interest in this paper.

The areas marked as dark spaces in Fig. 1 are not to be considered as devoid of all light. As shown in the intensity profile, the light becomes considerably less intense but never to blackness. Light emitted by the positive column is characteristic of the gas in the discharge and of the excitation conditions. Air is a salmon color (although the negative glow is blue), Argon is reddish purple and Neon is very intensely red-orange. Color was probably the key factor in selecting Neon for the very early Neon advertising signs.

The glow discharge discussed above is upset sometimes by the existence of striations (Fig. 2). These are defined as regions of intense light bordered by relative darkness; the entirety of which are contained in the positive column. This definition does not include the various dark spaces and glows associated with the cathode or anode. Striations fall into two main classes and two sub-classes. The main classes are "standing" and "moving" depending entirely upon the state of motion of the striation. The sub-classes of moving striations are "positive" and "negative"; positive being those which travel from anode to cathode while the negative striations move just the other way.

Standing striations appear to be fuzzy spherical regions of light whose diameter is slightly less than the diameter of



STRIATED DISGHARGES

FIGURE - 2

the tube. Under certain conditions they will assume a part-spherical shape so that when viewed through the side of the tube they appear crescent shaped (Fig. 2-b). Cylindrical and other shapes are unknown.

Moving striations are not generally seen with the naked eye but are viewed with a rotating mirror or an oscilloscope-photomultiplier tube combination. A typical oscilloscope trace is sketched in Fig. 3. Moving positive striations are typically repetitive at 800 to 1500 $sec.^{-1}$ with velocities of 60 to 100 meters per second and longitudinal separations of five to fifteen centimeters. Negative striations are less commonly encountered, more difficult to detect and of no interest in this work. No further description of these will be given.

It is to be noted here that the unstriated glow discharge is an excellent vehicle for the study of plasma properties and that better understanding of striations may lead to a simple scheme for suppressing them. If so, then we shall have the very much sought "stable homogeneous plasma".

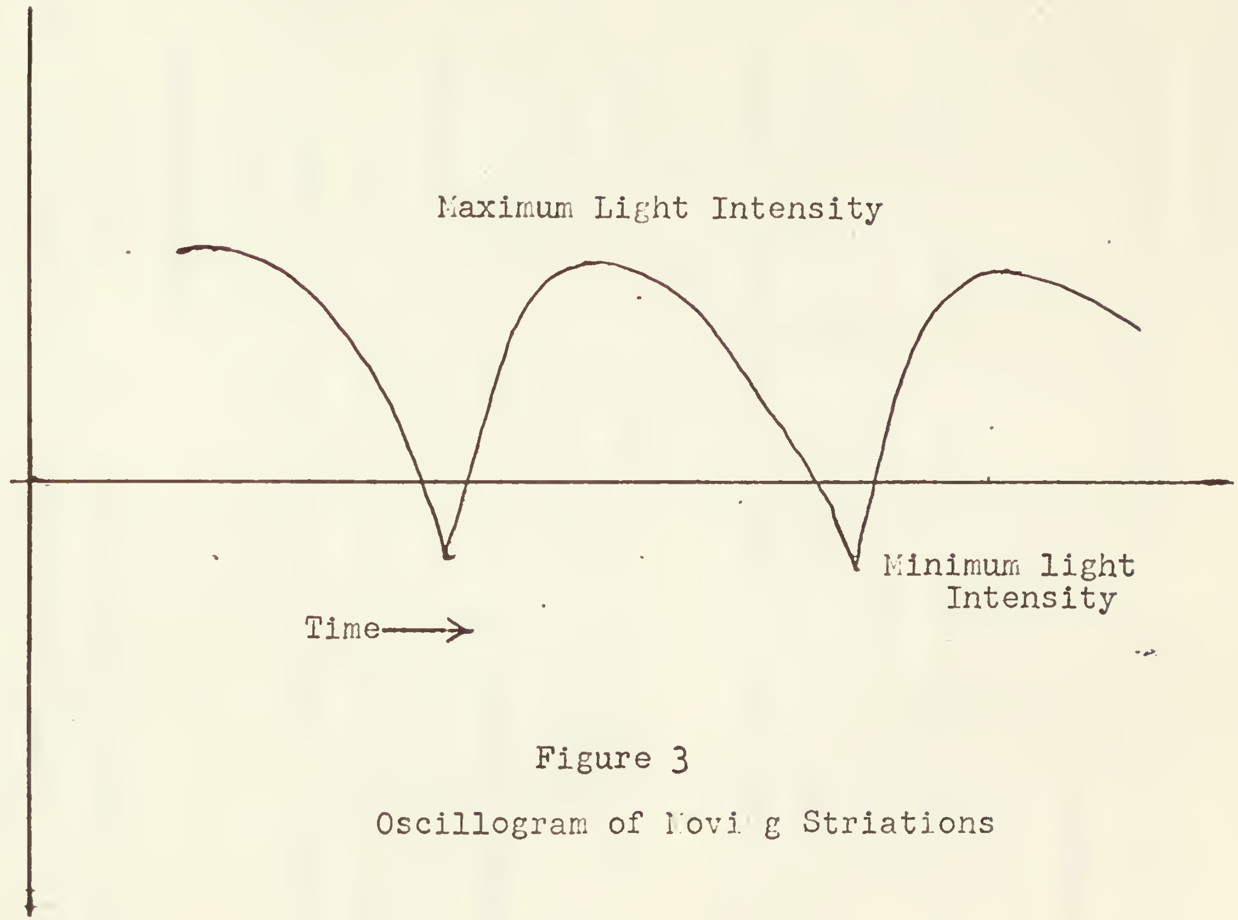


Figure 3

Oscillogram of Moving Striations

2. EXPERIMENTAL EQUIPMENT

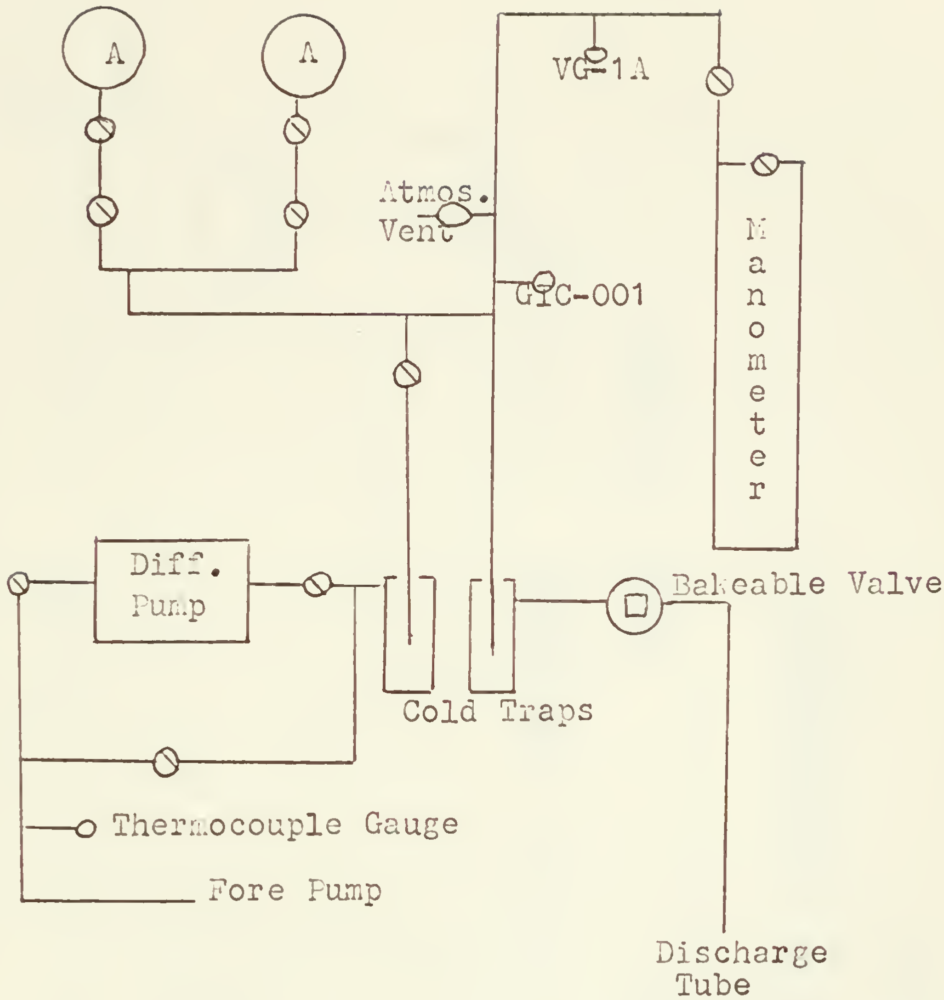
2.1 Vacuum System

The vacuum system sketched in Fig. 4 was designed and constructed by A. W. Cooper in 1959. It is composed, in the main, of a Welch fore pump, a Consolidated Vacuum Corporation oil diffusion pump, a bakeable high vacuum valve and two gas bottles. The entire assembly is plagued with large volumes and sharp corners but offers the advantages of quick tube changing and bakeout; advantages much needed by the many neophyte researchers who have used them.

Pressures in the system below one micron were measured by Consolidated Electrodynamics Corporation ionization gauge type GIC-100 coupled to the CEC sensing element GIC-001. Pressures above one-tenth millimeter Mercury were measured by a U-shaped manometer filled with Octoil-S (one meter Octoil-S equals 6.72 centimeters Mercury). Pressures between these ranges were of no interest.

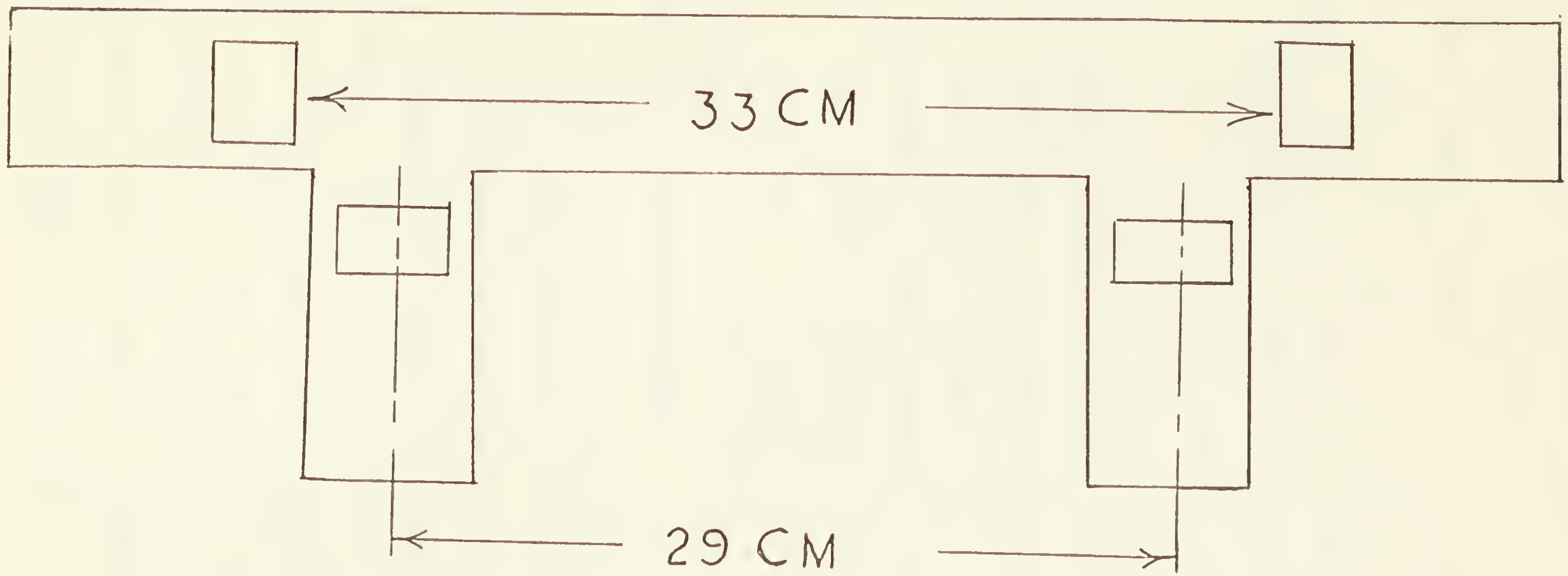
2.2 Discharge Tubes

In the course of this investigation several discharge tubes were constructed for several purposes. The main discharge tube is sketched in Fig. 5. The side arm electrodes of this tube were used during all observations and the side arms themselves were blackened well with soot. The use of Pupp's anodes in all electrode assemblies is to permit use of either end of the tube as cathode, thereby permitting nearly uninterrupted operation in the event of cathode burnout.

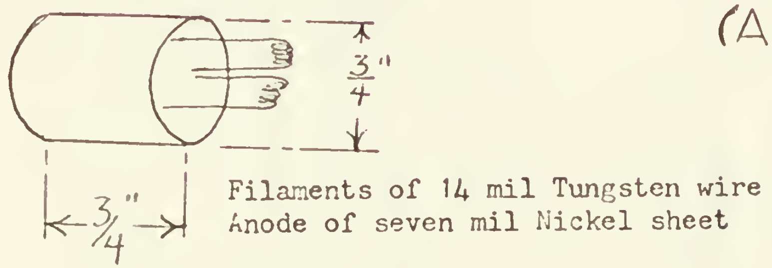


The Vacuum System

Figure 4



(A) TUBE

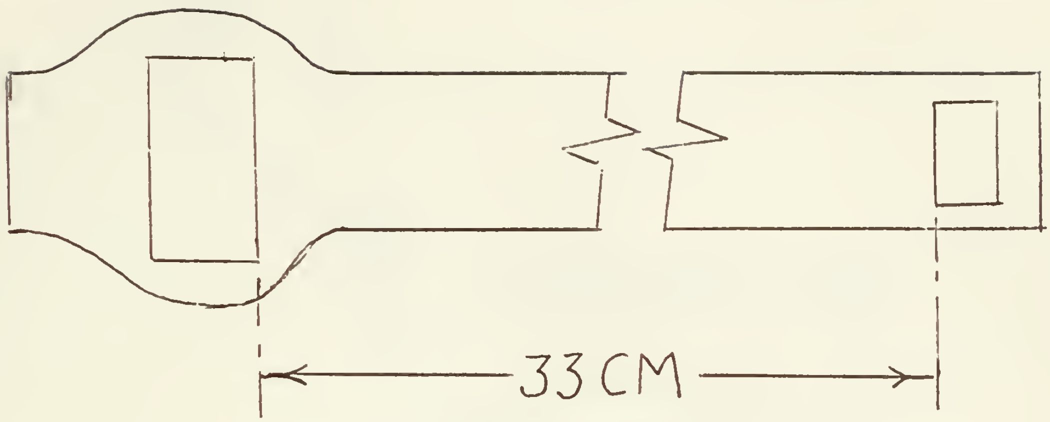


(B) ELECTRODE

FIGURE - 5

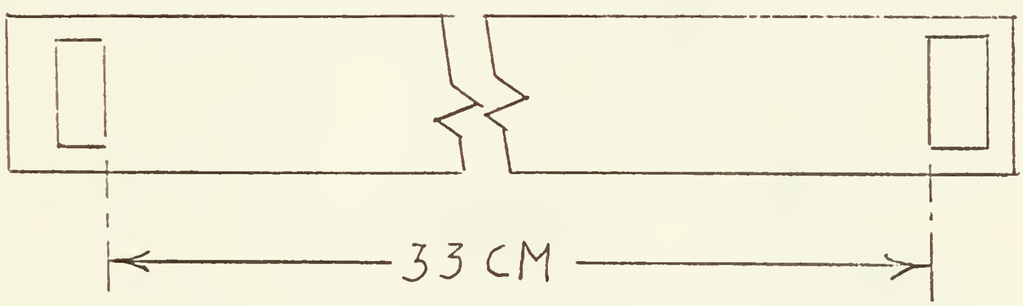
The upper and lower irradiating tubes are shown in Figures 6 and 7. The lower tube is a straight tube with simple wound filaments so constructed as to permit very high currents. The upper tube, also straight, differs from the lower in that one electrode assembly is formed with an oxide coated cathode. The very copious electron emission from such a cathode forms a cloud of electrons around the cathode and hence forms a virtual cathode at the cloud. This virtual cathode prevents the very high damage and heating usually experienced when wound tungsten cathodes are subjected to large scale positive ion bombardment. Neither of the irradiating tubes were configured for Rupp's anode operation and both were operated at above the critical current for striations. In neither tube did the anode oscillations extend sufficiently far into the positive column to interfere with the experiment.

One other irradiating tube was constructed as a part of this investigation. This tube, sketched in Fig. 8, was to be literally wrapped around the main discharge tube as suggested by Lischke and Schmidt,[21] to provide localized illumination of that discharge. A full explanation of the purpose of this tube will be given in section 3.2.3. Suffice it to say that initiating a dc discharge in such a tube configuration is difficult and that once begun, the discharge could not be operated at the very high current required for intense illumination.



Upper Discharge Tube

Figure 6



Lower Discharge Tube

Figure 7

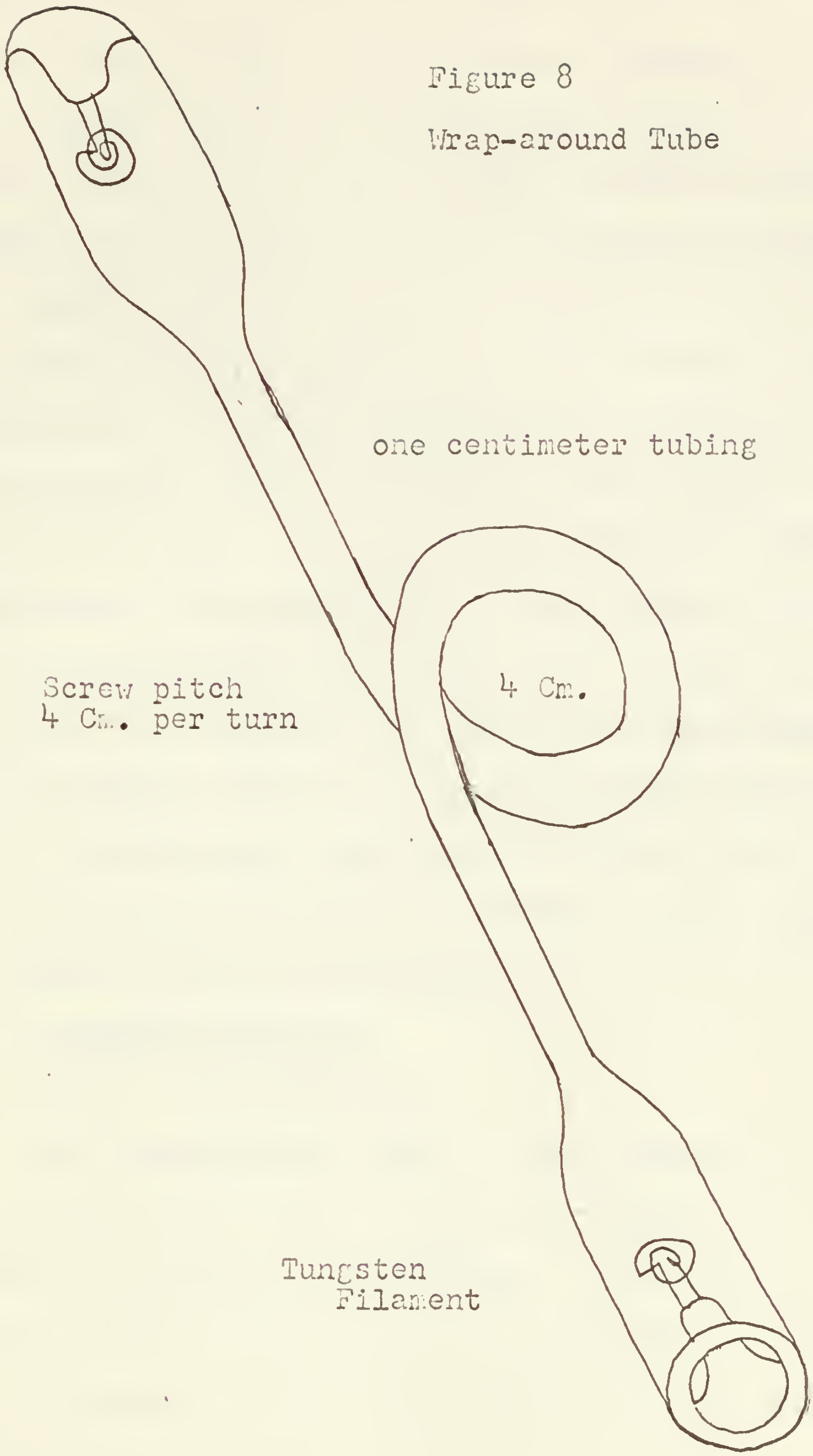


Figure 8

Wrap-around Tube

Since clean tubes and pure discharges go hand in hand, the following procedure was used to purge the tubes as well as could be done. Each tube was attached to a separate pumping system and placed under vacuum for 12 or more hours. While still under vacuum the anode strips were heated by induction and left a bright red for several minutes. During this same time the filaments were heated to a brilliant white by applying 60 cycle current to them. After cleaning the electrode assemblies, the tube was wrapped with Electrothermal tape and allowed to bake under vacuum for 24 hours or more. As a final step before transfer to the experimental vacuum system, an Argon discharge was ignited in the tube and the tube pumped until the discharge was extinguished. On completing three such discharge-pump cycles the tube was filled with Argon to about one-third atmospheric pressure and then vented through a Drierite bottle to atmospheric pressure. Transfer to the experimental system was now accomplished.

2.3 OPTICAL EQUIPMENT

2.3.1 Monochromator

The monochromator used in this investigation is a Baird Associates one meter grating monochromator fitted with a Baird Associates beam splitter. The grating is blazed for maximum transmission in the 35,000A to 45,000A region corresponding to the fifth order of diffraction of the wavelengths of interest. Slits of 25 micron and 75

micron width were available. The 75 micron slits were chosen to provide greater illumination to the photomultiplier tube fitted to the output of the monochromator. The 75 micron slits are capable of resolution of $\pm$ 3Å. Such resolution is nearly the limit obtainable on the monochromator so that no meaningful increase in selectivity could be gotten with narrower slits.

The monochromator is fitted with a 4-dial counter as an arbitrary means of positioning the grating. It was determined that the monochromator so fitted has wavelength as a linear function of dial reading, the relationship being

The selectivity is then

$$\frac{d\lambda}{dD} = 30 \text{ Å/Div.}$$

or more precisely to account for the order of diffraction,

$$\frac{d\lambda}{dD} = \frac{30}{N} \text{ Å/Div.}$$

Since the dial could be read only to the whole unit, the maximum selectivity in the fifth order is then,

$$\frac{d\lambda}{dD} = \frac{30}{5}$$

$$\frac{d\lambda}{dD} = \pm 3 \text{ Å/Div}$$

2.3.2 Photomultiplier Tube

Undoubtedly the most critical of all equipments used was the photomultiplier tube which detected outputs from the monochromator. Physically, the PM tube was mounted on the beam splitter at the output slit of the monochromator as shown in Fig. 9. The beam splitter is so configured that the case which enclosed the PM tube could be screwed onto the beam splitter. The PM tube used was the RCA 7102 selected because its spectral sensitivity is maximized in the region of interest (see Fig. 10).

The high voltage to the PM tube came at first from a high voltage electronic supply and later from a group of three 300 volt dry batteries connected in series. As had been suspected, the electronic power supply ripple voltage rendered the PM tube unusable at the low intensities available from the main discharge tube. With the battery pack, the sensitivity of the PM tube is limited only by the dark current noise and the light noise. The dark current noise is due to thermionic emission of electrons from the photocathode when the photocathode is at temperatures above zero degrees Kelvin. Light noise is the random release of electrons from the photocathode due to incident photons and is proportional to the square root of light intensity. The dark current noise can be lowered by refrigerating the PM tube but there is no way of reducing light noise given that the photocathode receives some illumination.

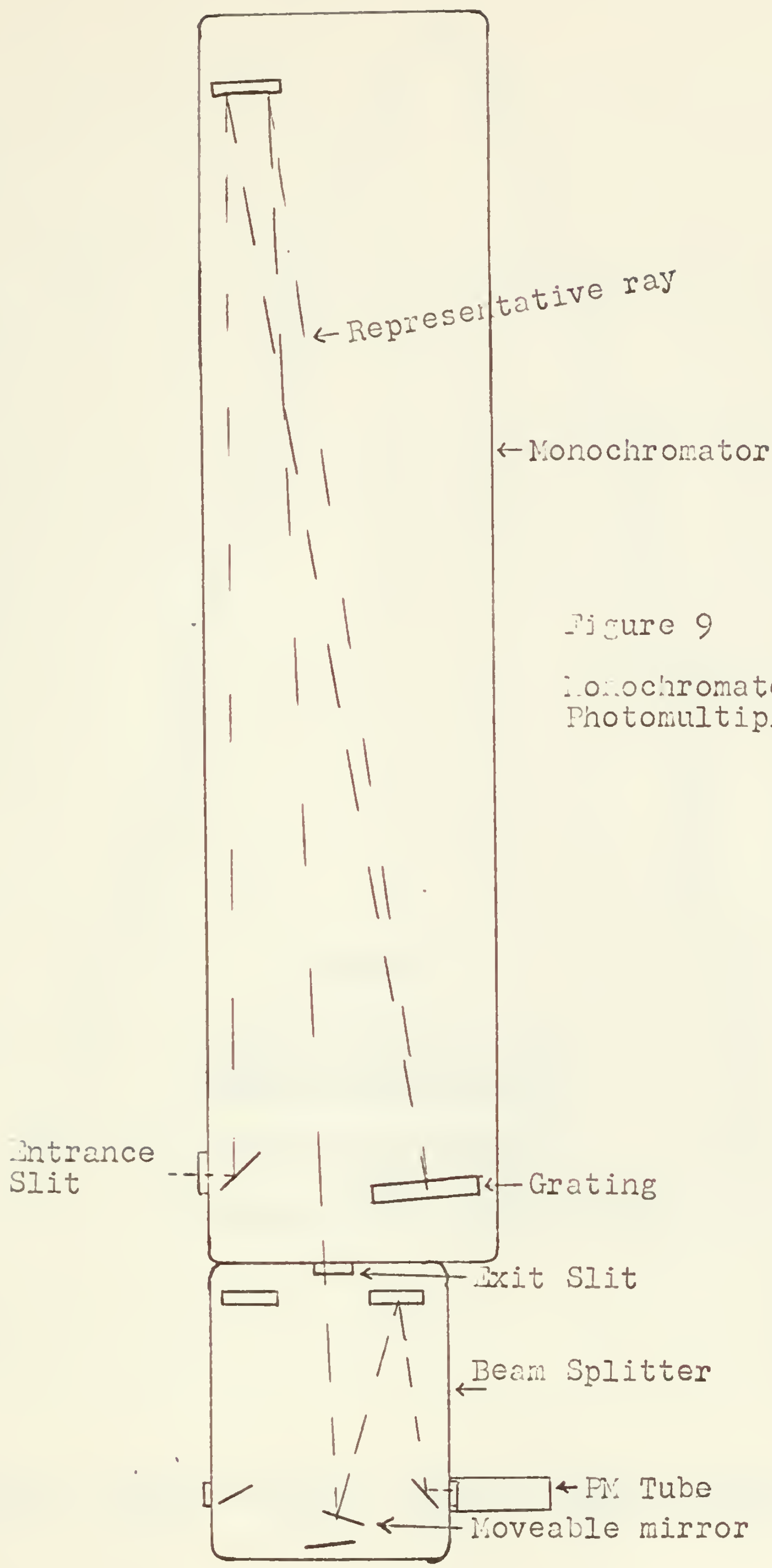


Figure 9

Monochromator and
Photomultiplier Tube

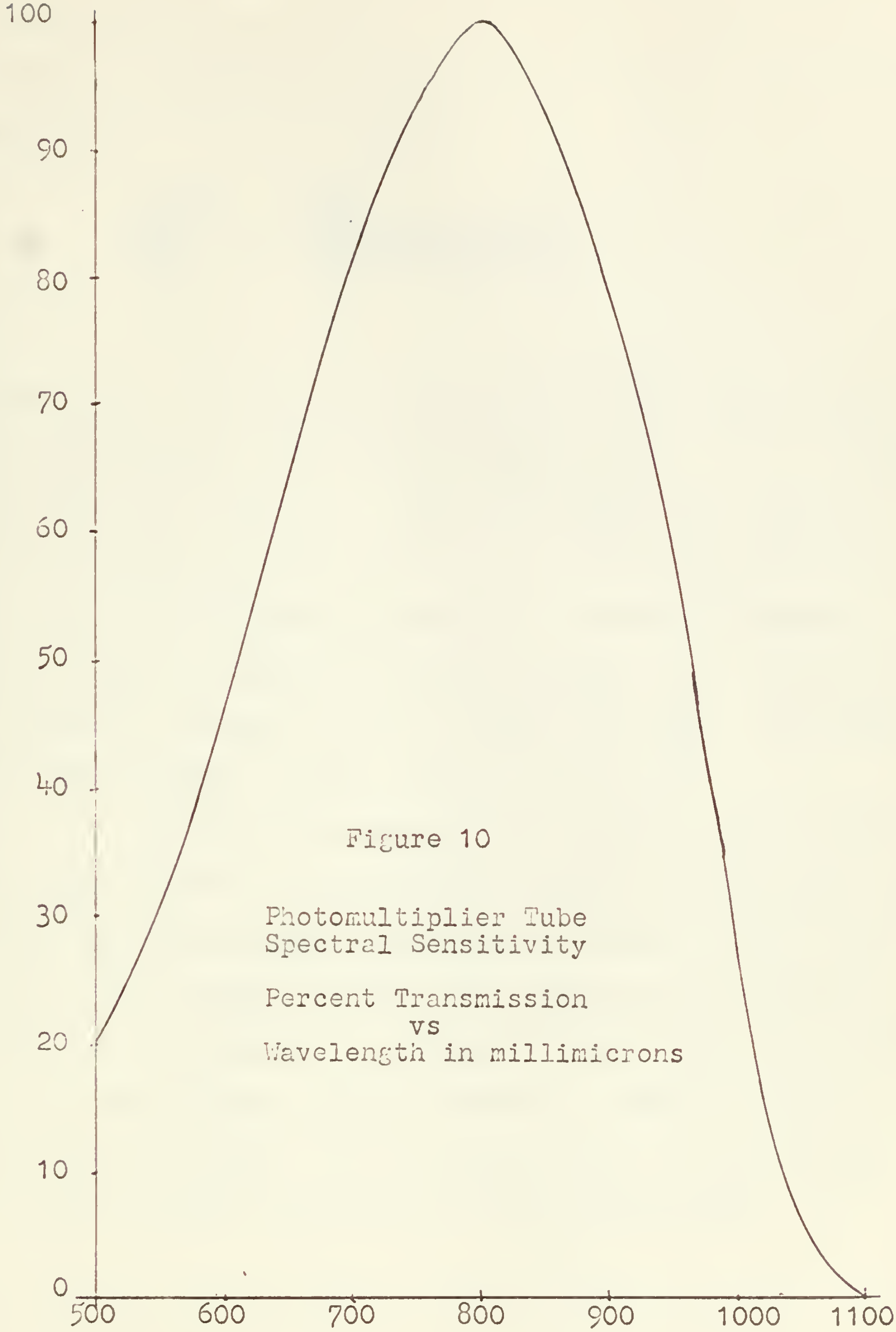


Figure 10

Photomultiplier Tube
Spectral Sensitivity

Percent Transmission
vs
Wavelength in millimicrons

The signal to noise ratio is of great interest in this experiment because of the very low intensities involved. The signal to noise ratio is related to the photocathode current by the equation

$$\left(\frac{S}{N}\right)^2 = \frac{(i_1)^2}{2e\,\Delta f(i_1+i_d)}$$

where i_1 is the light current, i_d is the dark current, Δf is the bandwidth and e is the electronic charge. It is readily seen that for $i_1 \gg i_d$ the signal to noise ratio increases with the square root of the light current which increases, in turn, with the square root of the incident intensity. Accordingly, to double the signal to noise ratio requires a fifteenfold increase in the incident intensity. This point will be discussed in more detail in section 4.

In order to lower the dark current, the PM tube was inserted into a refrigerator container in an effort to reduce the photocathode temperature. Details of the container are contained in Fig. 11. Although the container could not be evacuated as desired, it did reduce the photocathode dark current below a detectable level using 900 volts on the PM tube and the maximum amplification setting of 0.05 volts per centimeter on the oscilloscope.

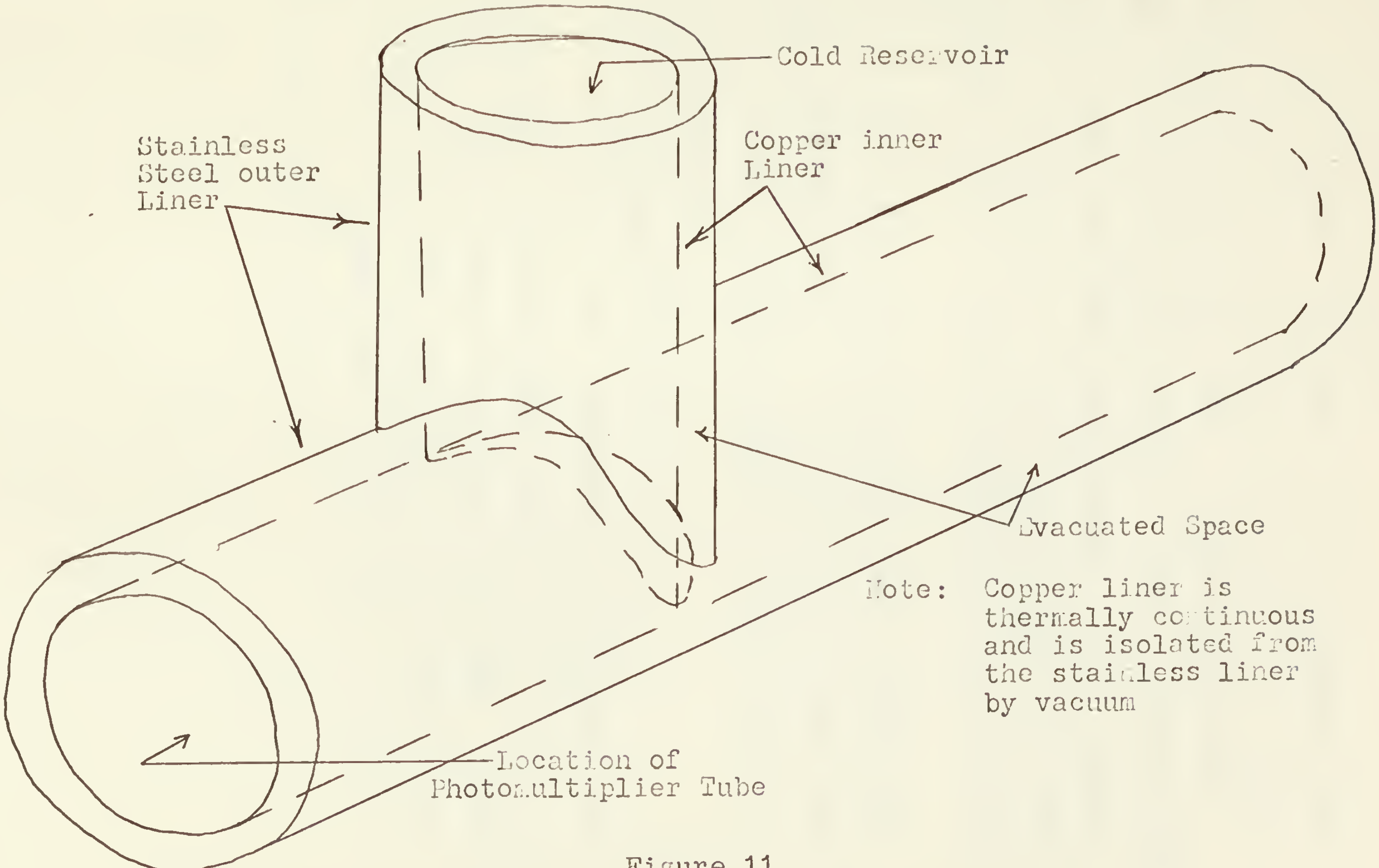

Cold Reservoir
Stainless
Steel outer
Liner
Copper inner
Liner
Evacuated Space
Note: Copper liner is
thermally continuous
and is isolated from
the stainless liner
by vacuum
Location of
Photomultiplier Tube

Figure 11

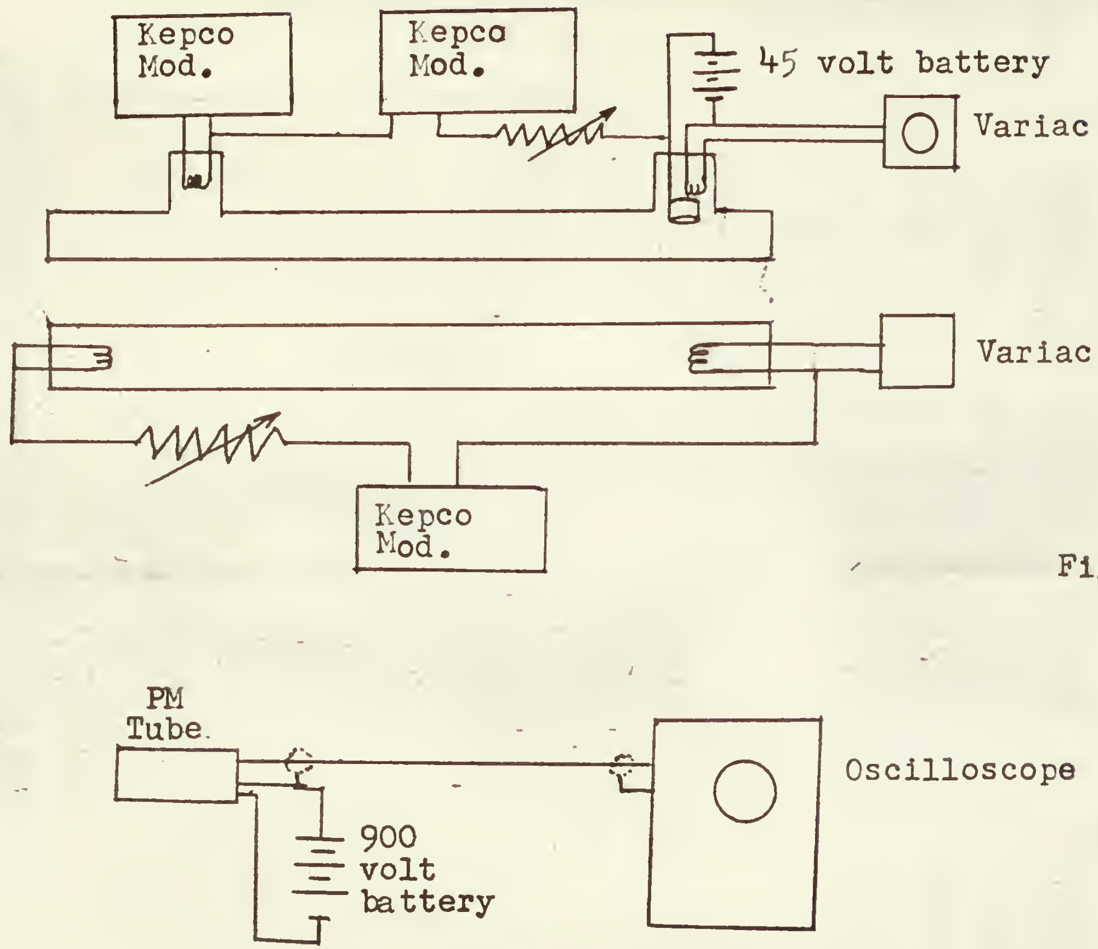
2.4 Electronic Equipment

The circuit schematics for the discharge tubes and the PM tube are shown in Fig. 12. The tube ballasts shown were all high current (500 to 1500 milliampere) wire wound resistors. Those shown as variable were either slide wire (for below 500 milliamperes) or heavy rheostat type. The oscilloscope is the tektronix 536 scope which admits plug in units for both the horizontal and vertical sweeps.

All voltages except those of the PM tube were measured by VTVM with high (11 megohm) input impedance and all currents were measured by Weston current meters of appropriate range.

2.5 Miscellaneous equipment

Throughout the investigation, sundry spectrum tubes, clamps, a spectroscope, various sorts of lenses and mirrors and other pieces of equipment were used. These were obtained from the Optics laboratory of the Postgraduate School and are commonly found in such labs. No further description need be given.



Electronic
Circuit
Schematics

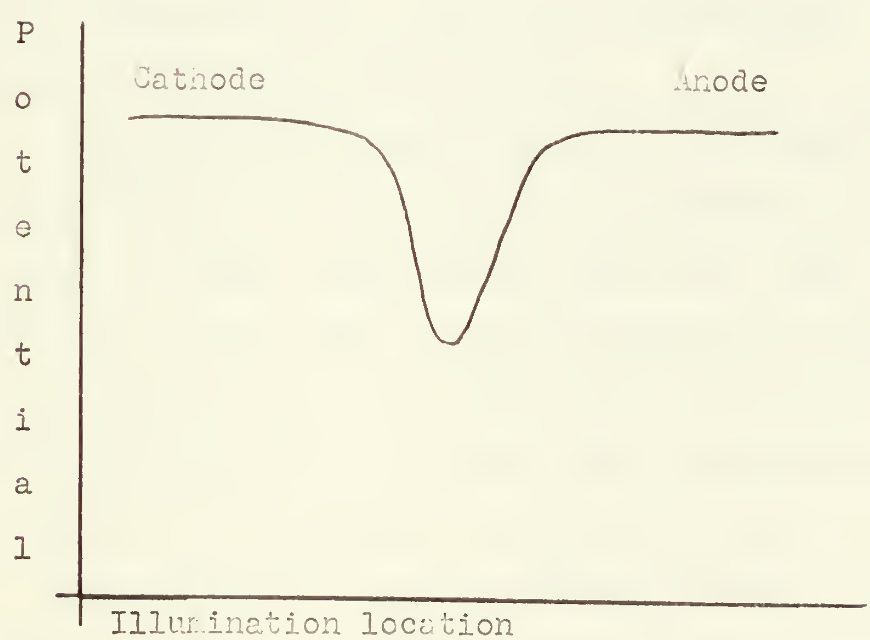
Figure 12

3. INVESTIGATIVE EFFORTS

The significant investigative efforts were three in number, two in direct support of this paper and the third to attempt to corroborate a phenomenon described by Robertson. These will be discussed in reverse order.

3.1 Discharge Tube Potential

In private communication to A. W. Cooper, Robertson has described a phenomenon observed by him in glow discharges. He arranged to illuminate only a narrow section of a discharge with light from a second discharge and he made the location of such illumination along the positive column a variable. He noted that the potential difference across the illuminated tube varied somewhat in the manner shown



Since the wrap-around tube was available for localized illumination, Cooper suggested an attempt to corroborate the existence of this phenomenon. Several attempts were made with the illuminated main discharge running at carious currents below 50 milliamperes but no comparable changes in tube potential could be obtained.

Since the wrap-around tube had to be run on ac rather than on dc and since the sources of ac available were of rather low current capacity it is suspected that the lack of results is due to a lack of intensity of illumination. No attempts were made to run the wrap-around tube on dc for which high current sources could be gotten since it had been earlier determined that potentials in excess of 1200 volts dc were required and these were not conveniently available.

3.2 Determination of Metastable Populations

3.2.1 Theoretical Introduction

The theoretical treatment given to striations by Robertson[14] does, by that author's own admission, suffer from the use of small perturbation theory and lack of dependable data on the time rate of ionization and excitation of the discharge atoms. In a later work co-authored with Hakeem[18] in 1961 Robertson concludes that the dependence of striations on populations of metastable levels is so clearly demonstrated that a more polished theory might be worthwhile to attempt. It was to this end that Mishcke and

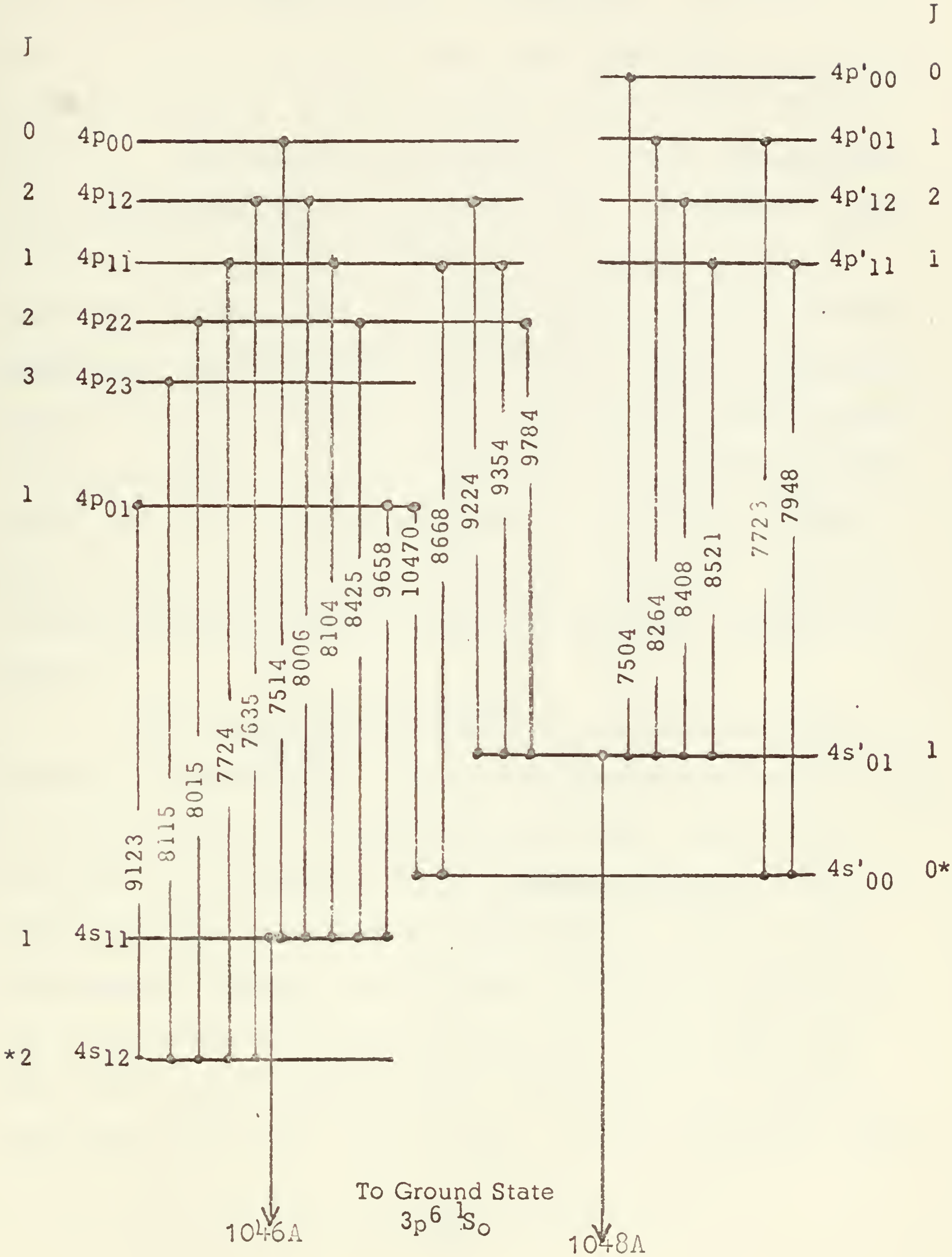
and Schmidt[21] attempted to determine metastable populations in 1962. This very same determination was attempted in this investigation.

There are basically two techniques for determining the populations of a level. Either one observes the amount of light emitted by atoms when transiting from that level to some lower level (for example, the ground state) or one observes the attenuation of a beam of radiation as it passes through the region where atoms are populating some level. The attenuation is then a measure of the number of transitions out of the level and hence is a measure of the population of that level. Both techniques were attempted as part of this investigation.

Fig. 14 shows some of the transitions associated with the metastable levels in Argon. As can be seen, irradiating with the single wavelength 8115A induces a transition from the metastable $4S_{12}$ level to the $4P_{23}$ level from which level there is only one possible path, namely back to the $4S_{12}$ level. Accordingly, in the presence of radiation, there will be a momentary disturbance of the level populations but these will quickly return to their former values. Such a transition which does not disturb the populations should not disturb the striations if striations are strongly dependent on some mechanism directly involving the metastable levels. It is for this reason that 8115A is a desirable wavelength at which to work.

Fig. 14

Optical Transitions Associated with the Argon I Metastable Levels. AIP Notation



3.2.2 Emission Technique

Mischke's and Schmidt's technique for measuring the effect of radiation on striations is basically sound. Their scheme as shown in Fig. 15 was simply to observe two locations in the discharge which were separated by one striation wavelength. The two PM tube signals were then cancelled in a differential preamplifier and the preamplifier output was displayed on an oscilloscope. By balancing the PM tube signals exactly with no irradiation and then irradiating at one of the observation points any additional emission of the interesting wavelengths could be determined since the PM tube signals at the differential preamplifiers would be different. This scheme requires matched monochromators and nearly matched PM tubes. Since only a single monochromator was available and their efforts to construct a second met with failure, they were unable to report any results.

The approach here differs in several ways from Mischke's and Schmidt's. In this investigation (see Fig. 16), the main discharge is illuminated by a very high current illuminating discharge through a narrow bandpass filter. Having only one monochromator there was no need for the differential preamplifier or second PM tube. The PM tube was refrigerated for this experiment to reduce dark current noise and the monochromator was fitted with narrower (75 micron vice 750 micron) slits. It was anticipated that

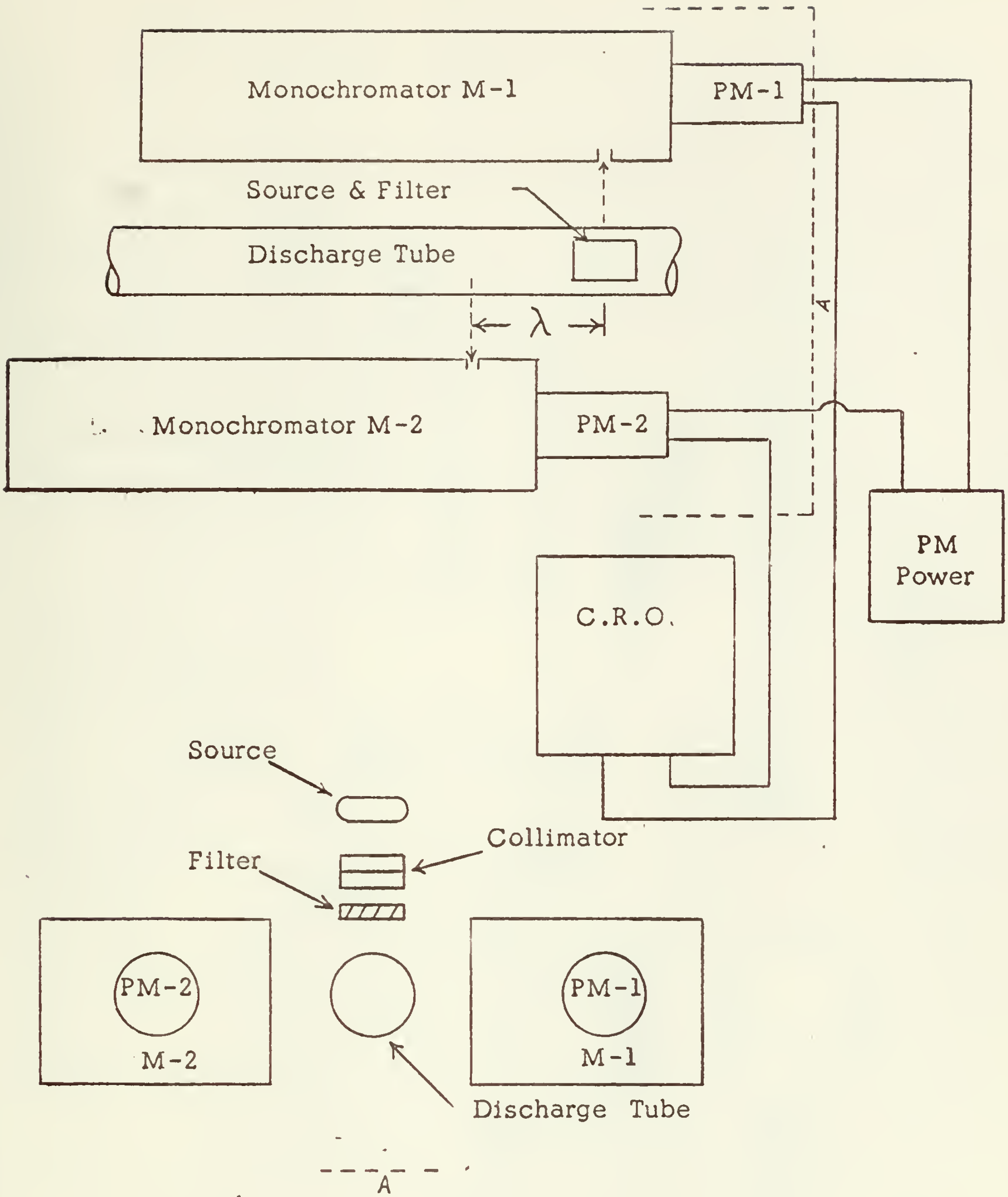
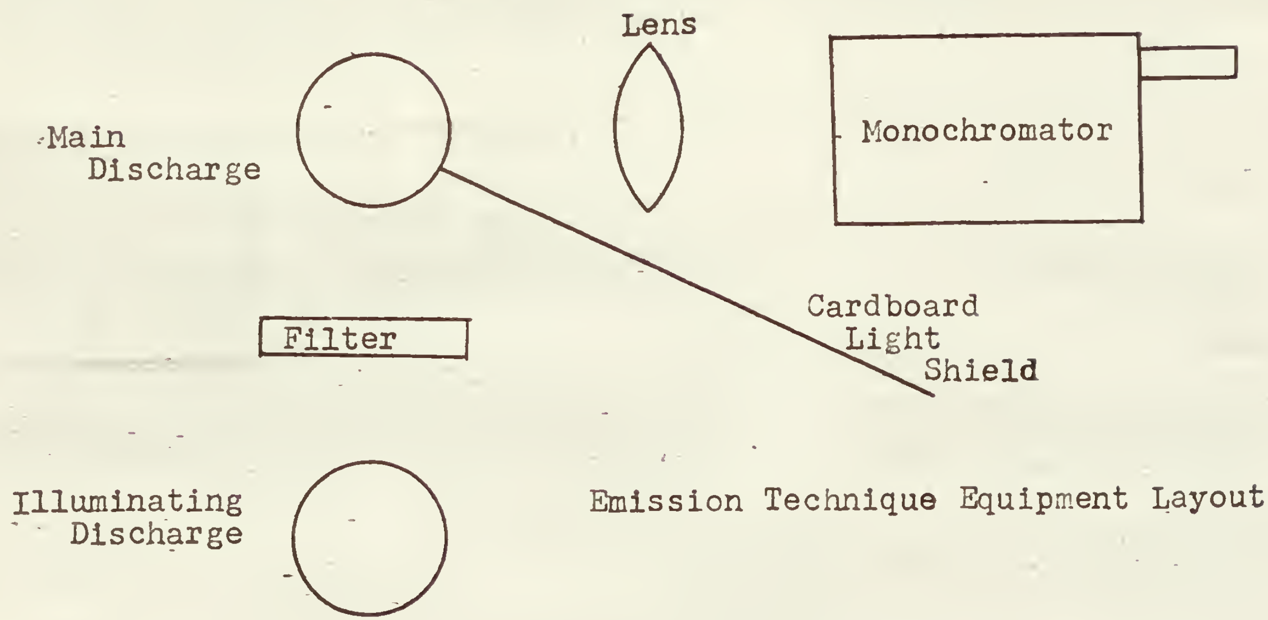


Fig. 15. Equipment Arrangement for the Emission Technique of Mischke and Schmidt



Emission Technique Equipment Layout

Figure 16

with narrower slits, the cold PM tube and a battery pack for the PM tube, the very tiny changes which should occur in the emission of the 8115A line could be detected.

In the experiment, the main discharge was run at various currents from 150 milliamperes (the lower limit of phototube detection) to the power supply maximum of 500 milliamperes and at pressures from 0.33 millimeters Mercury to 3.4 millimeters Mercury. The illuminating discharge was operated above the critical current for striations (about 1200 milliamperes). The results of this phase of the work are examined in section 4.

3.2.3 Absorption Technique

In the technique used by Mischke & Schmidt one sees a sound idea poorly applied. They arrange two PM tubes (see figure 17) on orthogonal axes, both tubes observing the striated discharge. The signals from these PM tubes are cancelled in a differential preamplifier. They then irradiate the discharge along the axis of one of the PM tubes and observe whether or not the resulting imbalance of PM tube signals is constant or modulated. They irradiate with a single wavelength selected by the monochromator. If the imbalance were constant this would indicate that the passage of a striation had no effect on the rate of absorption of radiation. If the imbalance were modulated, its relative waveshape and phase relationship to the striation could be used to obtain information on the time rate change

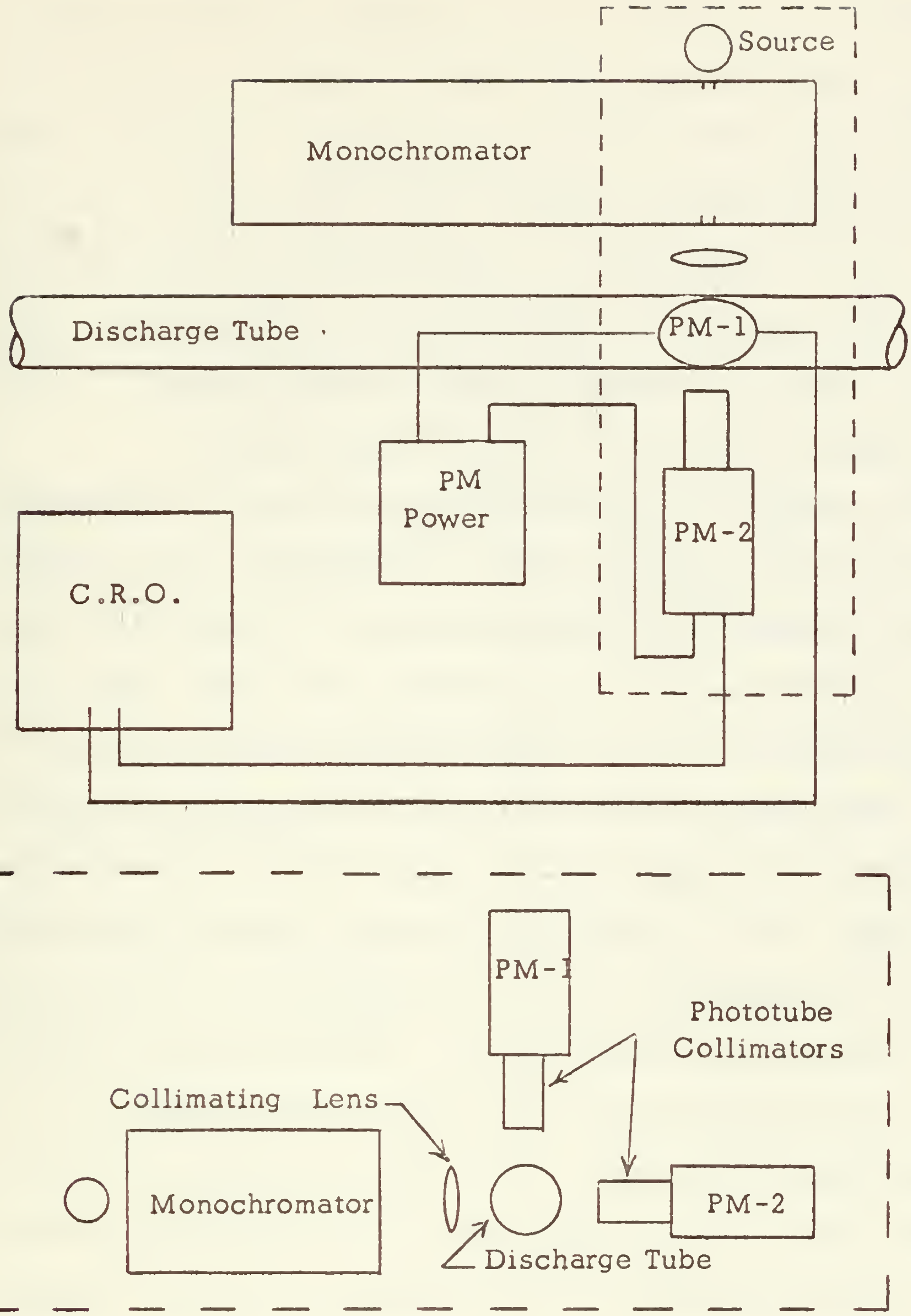
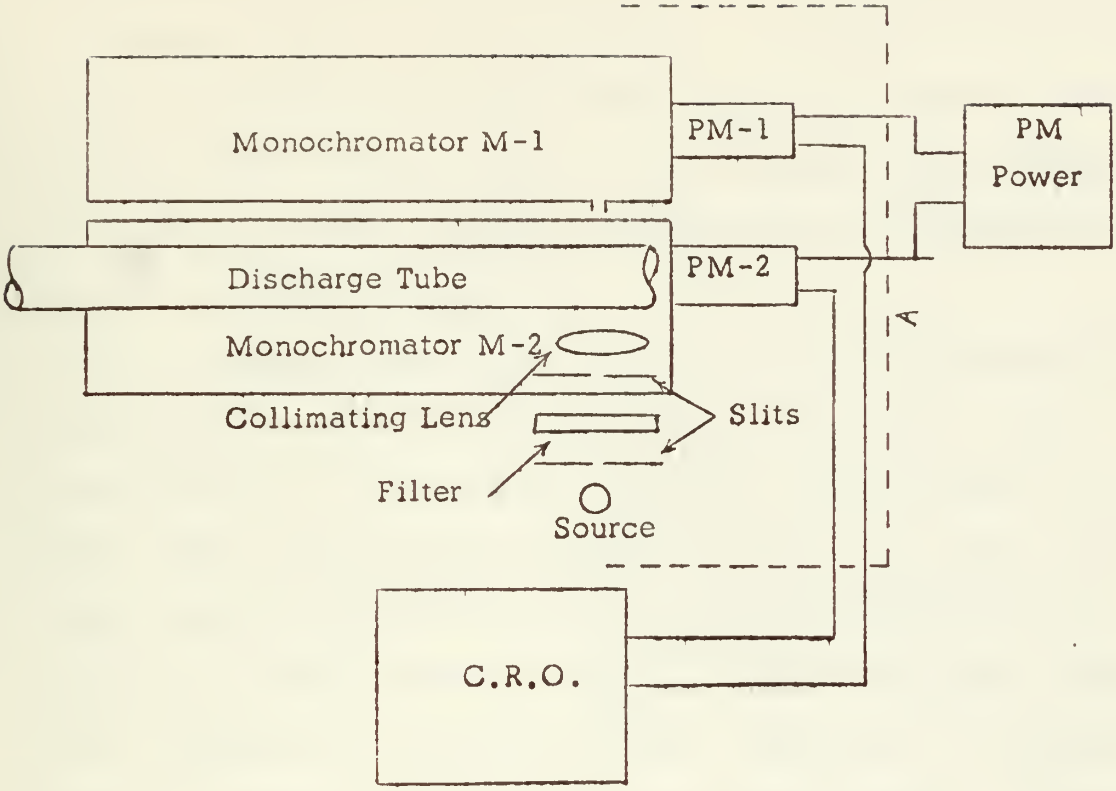


Fig. 17 Equipment Arrangement for the Absorption Technique of Mischke and Schmidt

of population of a metastable level. Since the technique produced no conclusive results, they suggest an improved absorption technique which involves two matched monochromators and PM tubes. The improved scheme, (see figure 18) appears to be the kind which will produce results but still requires the additional monochromator. Not having one and realizing that construction of a second is, at best, a risky investment of rather limited time, I chose to proceed differently.

Before embarking on my approach, a short time was invested in re-performing the Mischke and Schmidt version. Rather than a differential preamplifier, both PM tube signals were displayed on the oscilloscope and placed in superposition. The main discharge was then irradiated through a narrow bandpass filter peaked for 810 millimicrons but wide enough to permit the 8115A line to be transmitted. When irradiating, the PM tube oriented along the direction of radiation showed a sizeable increase in dc signal but no significant differences in the ac components of the PM tube signals could be seen. The difficulty was in the comparison of the scope traces since the trace displaying the radiation had to be placed on a much less sensitive amplification setting in order to keep the trace on the scope face. This caused a concurrent decrease in the ac component, rendering comparison of the two traces difficult at best. Using the ac only inputs to the scope did nothing to improve matters except to permit equal amplification of the two signals.



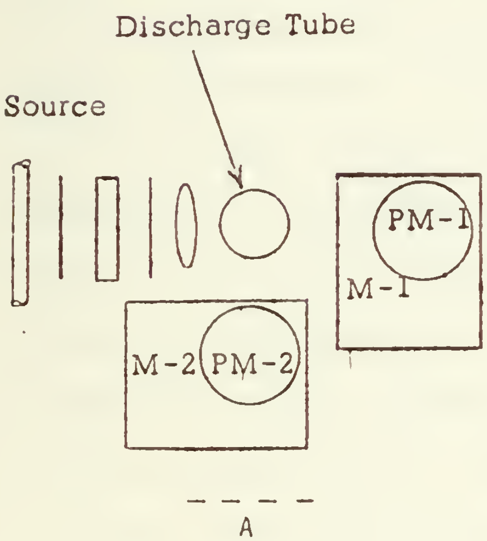


Fig. 18. Recommended Equipment
Arrangement for the Improved
Absorption Technique.

Comparison was again difficult because the light from the irradiating tube is not a constant. It fluctuates naturally and, in addition, shows 120 cycle modulation which stems either from the anode power supply or the filament power supply. Time did not permit further investigation or attempts to filter out unwanted components.

In the absorption part of the investigation the equipments are arrayed as shown in the figure 19. The monochromator was adjusted to maximum transmission in the fifth order of diffraction for the 8115A line first with an Argon spectrum tube and finally with the light from the irradiating tube passed through the narrow bandpass filter. After alignment and adjustment were completed the irradiating tube was shielded and the main discharge ignited. Observation with the second PM tube indicated the existence of moving striations. As was expected, the monochromator and its detecting PM tube were unable to observe the striation waveform of the 8115A line because of the very low intensity of the main discharge (discharge currents here were below 50 milliamperes and above 10 milliamperes. The lower limit was selected to just prevent extinguishing the main discharge by the irradiating tube). It was not until the main discharge tube current reached nearly 150 milliamperes that the 8115A line showed through the monochromator.

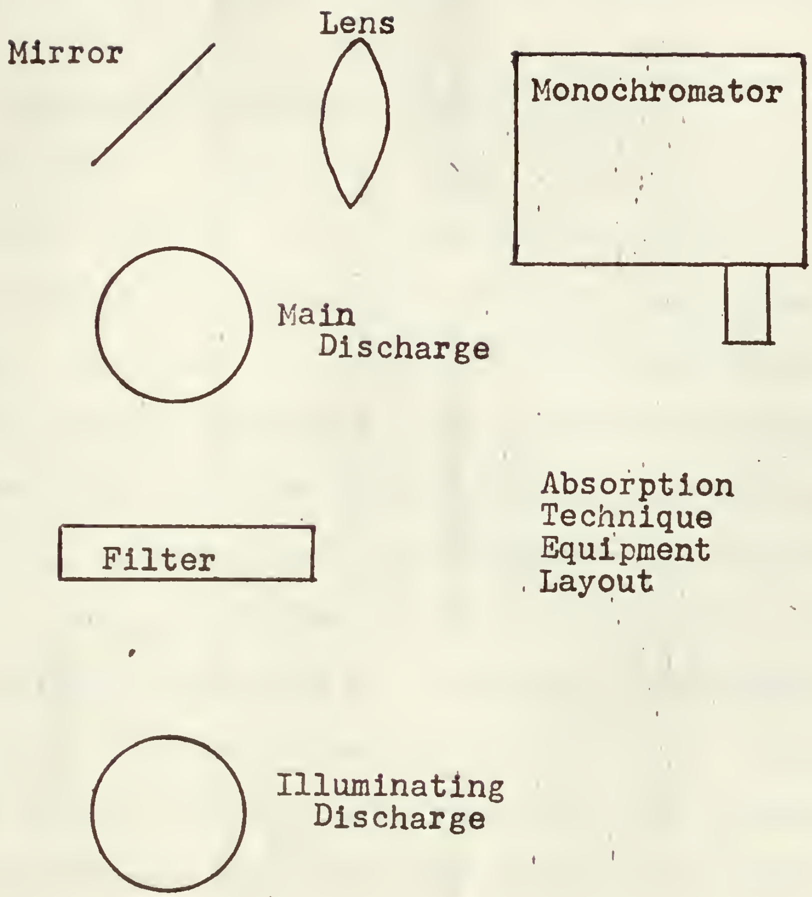

Lens
Mirror
Monochromator
Main
Discharge
Absorption
Technique
Equipment
Layout
Filter
Illuminating
Discharge

Figure 19

The actual experimental procedure at this point was to observe the illuminating discharge, through the main discharge, while the main discharge was extinguished and to observe again when the main discharge was running. The second PM tube signal was displayed throughout the observations. This procedure was repeated for various main discharge currents from ten to 100 milliamperes and pressures from 0.33 to 3.4 millimeters Mercury.

The results of this part of the experiment are discussed in Section 4.

3.2.4 Additional Emission Investigation

As shown in figure 20, there are several population and depopulation schemes. The inconclusive results of the absorption and emission techniques could be interpreted in two ways, viz. that there was insufficient illumination from the illuminating discharge in the desired wavelengths or that the entire experimental philosophy suffers from some undetected flaw. In an effort to establish one of these possibilities, the main discharge was irradiated as called for in the emission technique but different wavelengths were selected by the monochromator for detection. Irradiating with the filter for the 8115A line permitted passage of the 8104A line as well because of the bandpass of the filter. Using this filter, a search was made for an increased emission of the 9354A line by which the $4P_{11}$ level transitions to the $4S'_{01}$ level and thence to the ground state. Using a filter

Transitions Associated with Argon Metastable Levels

$4S^*_{12}$	8015A	$4P_{22}$	8425A	$4S_{11}$	to ground state
			9784A	$4S'_{01}$	to ground state
$4S^*_{12}$	8115A	$4P_{23}$	8115A	$4S^*_{12}$	
$4S_{11}$	8104A	$4P_{11}$	7724A	$4S^*_{12}$	
			8668A	$4S'^*_{00}$	
			9354A	$4S'_{01}$	to ground state
$4S^*_{12}$	7724A	$4P_{11}$	8104A	$4S^*_{11}$	
			8668A	$4S'^*_{00}$	
			9354A	$4S'_{01}$	to ground state
$4S'^*_{00}$	7723A	$4P'_{01}$	8264A	$4S'_{01}$	to ground state
$4S_{11}$	8006A	$4P_{12}$	7635A	$4S^*_{12}$	
			9224A	$4S'_{01}$	to ground state

Figure 20

centered at 8015A and passing 8006A as well a search was made for increased emission of the 9784A line, the 8425A line and the 9224A line all of which permit 4P levels to proceed via 4S levels to the ground state. Finally with a filter which passes 7723A and 7724A a search was made for an increase in the 9204A line and the 9354A line which permit depopulation of the $4P'_{01}$ and $4P_{11}$ levels respectively to 4S levels and thence to the ground state. This phase of the investigation was carried out at a main discharge pressure of 3.4 mm Mercury and at currents above 200 milliamperes. The results of this phase of the investigation are discussed in Section 4.

3.3 Results of Investigations

Although the emission technique was run at several pressures and at various values of discharge current, no significant alteration of the PM tube signal occurred when the illuminating discharge was alternately shielded and unshielded. No combination of pressure and main discharge current within the ranges quoted in the description of the emission technique was fruitful despite many attempts and frequent use of an Argon spectrum tube to ensure that the monochromator and PM tube combination were operating properly and the main discharge was focussed on the entrance slit. These same remarks apply for the additional investigation of level depopulations although the same ranges of pressures and currents do not apply.

The much hoped-for results of the absorption technique also did not materialize. A goodly portion of the actual investigative effort was allotted to this phase because the technique showed such promise. For reasons I shall attempt to explain in Section 4, the improved technique fell short of expectations.

4. ANALYSIS AND DISCUSSION

As was suggested in section 3.2,4, there exists either a lack of illumination in the desired wavelengths or some flaw in the experimental philosophy. It was to resolve this question that the additional emission investigation was conducted.

It is this author's opinion that the experimental philosophy is sound and that the irradiating discharge simply failed to produce sufficient illumination in the desired wavelengths. The essential ingredient in this investigation is an intense source of 8115 Å radiation. This requirement stopped Mischke and Schmidt after they had tried black bodies and banks of spectrum tubes. This author's attempts to use a second discharge have come to naught and it appears that the source of radiation is again at fault. This time it might be easily corrected.

In developing the analysis here it is well to review some experimental evidence. In private communication to this author, F. W. Crawford noted that striations did not exist in Neon buffered Mercury discharges and demonstrated this very well during the summer of 1963. Tube currents were relatively small, being confined to below 500 milliamperes and the mercury partial pressures were one to three microns. The pressure of the buffering gas was about 5 millimeters Mercury. Both Foulds and Donohue and Wiele (23) have reported striations in unbuffered Mercury discharges under current and pressure

conditions similar to Crawford's. Mischle and Schmidt[21] are among the many authors who have reported moving striations in pure noble gas discharges wherein the tube currents were relatively small. Cooper and Oleson[22] have reported on the dependence of striations upon tube current and have shown that for a given pressure and tube radius there exists a current above which striations do not exist. Finally, the work of Hakeem and Robertson[18] in the alkali vapors has, taken together with earlier work, led to the conclusion that striations could not exist unless the discharge atoms were possessed of low lying metastable levels.

It is well also to review the main points of the Robertson[15] theory. He suggesta that the product of the metastable concentration and a quantity labelled F/ N must be positive and large enough to offset diffusion and wall losses in order for striations to exist. The quantity F is the total ionization rate of the metastable atoms by electrons and the quantity N is the concentration of electrons. Robertson dwells at length on the differential part of the product but says little about the role of the metastable concentration except to note that the concentration decreases with increasing tube current.

This author chooses to interpret the experimental results listed above against the background of the Robertson theory as follows. In the pure Mercury discharges of Foulds and of Donohue and Dieke and in the noble gas discharges of

Nischke and Schmidt it is likely that the Robertson idea of multiplying the metastable concentration by the partial derivative of F by N would give good predictions of the existence of striations and, if data were available, might be able to predict Cooper and Cleson's critical currents. In Crawford's buffered discharges it is possible that the magnitude of F as a function of N may have been less than it would have been in a pure discharge but there appears no reason why the partial derivative of F by N should not be as positive in one case as in the other. It would seem then, that the lack of striations can be attributed to a low population of the metastable mercury levels. That this is a reasonable presumption is supported by the greatly increased frequency of collision of the otherwise long-lived metastable levels in the buffered discharge over that experienced in the unbuffered case.

In the 1961 paper Robertson[16] passes off the lack of striations at high currents (hence high values of N) by suggesting that F as a function of N reaches a peak and then drops off, rendering the partial derivative of F by N a negative quantity. He does allow for the reduction in metastable concentration with the increasing current but does not attribute any great significance to this reduction.

In view of the lack of results of this experiment, I have concluded that even with the very high current through the illuminating discharge the emission of 8115A photons was

very much lower than expected. This I attribute to a low concentration of atoms in the metastable condition. While it may have been true that F/ N in the illuminating discharge was negative, thereby destroying striations, a lack of 8115A photons can only be attributed to a low population of atoms in the metastable $4S_{12}$ state.

There is some evidence to indicate that a metastable level, despite its long lifetime, is not as preferentially populated in a high current discharge as it might be under lower current conditions. As von Engel[24] has pointed out, metastable levels are not, in general, populated from lower lying states. The work of Kopfermann and Ladenburg[25] in Neon indicates that the relative populations of excited levels are distributed according to the Boltzmann factor with kT set at the electron temperature of the discharge rather than at the discharge thermometric temperature. This means that higher levels are relatively more populated at the expense of the lower levels. Also, the high current discharge requires a greater fraction of Argon atoms in the ionized state which leaves a smaller fraction to be in excited states.

Several authors[5,6,9,21] have reported on the effects observed while irradiating a discharge. None has succeeded in making single-wavelength irradiations of any significance so that the effects to be expected of such a radiation technique are, at best, conjectural. It seems safe to say that irradiation which might depopulate a metastable level should

produce an increased tube potential difference and, if we are to accept the Robertson theory, some change in the striation characteristics. The tube potential was continually monitored during the experiment by a VTVM and a PM tube observed the discharge light emission characteristics. The author did not note any significant differences in either the VTVM reading nor in the light intensity profile of the striations when the main discharge was or was not irradiated by the high current illuminating discharge. This observation is consistent with the notion that there was a low population density of metastable states in the illuminating discharge. The author suggests that others working with illuminated discharges did as Meissner and Miller (9) and arranged the illumination to produce some maximum effect on the illuminated discharge. That they did not mention striations in their illuminating discharges is explained by the smallness of the tube radii which permits a low value of critical current for striations and, of course, by their not using any time-resolving measurement devices.

In the construction of and operation of the illuminating discharge tubes, consideration was given to the total density of Argon atoms, to maximizing the macroscopic light output and to attempting to subdue striations. It appears that prime consideration must, instead, be given to maximizing the number of 8115A photons.

Additional work is required to devise suitable sources before any meaningful accomplishments will be made in determining metastable populations. To this end, an examination of the magnitude of 8115A light output should be undertaken with the purpose of determining the discharge conditions and tube configuration which will produce a maximum intensity at the desired wavelength.

It is suggested that, at first, a single tube about one meter long with side arm electrode assemblies be used. By selecting out the 8115A line with a monochromator and PM tube combination, intelligent intensity measurements can be made. When the critical current region is reached, intensity measurements can be made in both the striated and unstriated sections of the positive column in an effort to determine the effect of striations on the 8115A intensity.

After suitable sources of radiation are constructed, the procedure of Mischke and Schmidt for emission studies and the procedure in this work for absorption studies can be begun in earnest.

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